

THE NUCLEAR MOMENT OF BARIUM

By

ALBIN N. BENSON

A DISSERTATION

SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE DEGREE OF DOCTOR OF PHILOSOPHY
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The Nuclear Moment of Barium

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New data have been obtained on the hyperfine structure of several lines of barium. These have been studied in the attempt to determine more definitely the nuclear moment of the odd isotopes of barium, reported as $2\frac{1}{2}$ by Kruger, Gibbs and Williams, and as $1\frac{1}{2}$ by Schüler and by Murakawa. Studies of the separation of the $6^2S_{\frac{1}{2}}$ and $7^2S_{\frac{1}{2}}$ components from the center of gravity, and of the spacing and patterns of the $5^3D_{\frac{3}{2}}$ and $6^3P_{\frac{3}{2}}$ terms all lead to the conclusion that $I = 1\frac{1}{2}$ is the correct value of nuclear moment.

IN spite of several researches on the subject, the value of the nuclear moment of the odd isotopes of barium remains in doubt. The various investigators differ both in experimental findings and in interpretation of the data. McLennan and Allen¹ first found hyperfine structure in a number of the arc and spark lines of barium, but their results have never been confirmed by other workers. Frisch² found all lines sharp. The first accurate observations were those of Ritschl and Sawyer³ who observed structure in several lines and published measurements on the resonance lines of Ba II, $6^2S_{\frac{1}{2}} - 6^2P_{\frac{1}{2}, 1}$, from which Schüler and Jones⁴ deduced $1\frac{1}{2}$ as the nuclear moment of the odd isotopes. However, from their own measurements on the same lines, Kruger, Gibbs, and Williams⁵ concluded that the moment is probably $2\frac{1}{2}$, while Murakawa,⁶ from

observations on these and other lines, considered $1\frac{1}{2}$ to be correct. Recent compilations of nuclear moments⁷⁻⁹ have given $2\frac{1}{2}$ as the moment of barium but have indicated that uncertainty exists.

The difficulty in the determination of the moment from hyperfine structure observations arises from the fact that barium is a mixture of several isotopes. The recent measurements of Sampson and Bleakney¹⁰ give for the isotopes and their percentage abundance:

mass number	130	132	134	135	136	137	138
percentage	0.16	0.015	1.72	5.7	8.5	10.8	73.1

The even isotopes thus make up 83.5 percent of the atoms and all observers are agreed that these even isotopes, in common with all observed even isotopes save N¹⁴, have no hyperfine structure. In all barium h.f.s. patterns, the five even isotopes fall together in a very heavy central

¹ McLennan and Allen, *Phil. Mag.* **8**, 515 (1929).² Frisch, *Zeits. f. Physik* **68**, 758 (1931).³ Ritschl and Sawyer, *Zeits. f. Physik* **72**, 36 (1931).⁴ Kallman and Schüler, *Ergebn. d. Exakt. Naturwiss.* **11**, 134 (1932).⁵ Kruger, Gibbs, and Williams, *Phys. Rev.* **41**, 322 (1932).⁶ Murakawa, *Sci. Papers Tokyo I. P. C. R.* **18**, 304 (1932).⁷ White, *Introduction to Atomic Spectra* (McGraw Hill, 1934), p. 372.⁸ Darrow, *Bell Tech. J.* **14**, 319 (1935).⁹ Bacher and Bethe, *Rev. Mod. Phys.* **8**, 82 (1936).¹⁰ Sampson and Bleakney, *Phys. Rev.* **50**, 456 (1936).

TABLE I. Observed hyperfine structure in barium.

	CLASSIFICATION	PLATE SEPARATIONS	POSITIVE COMPONENT	NEGATIVE COMPONENT
4554.0	Ba II $6^2S_{1/2}-6^2P_{1/2}$	10	0.152 cm ⁻¹	0.100 cm ⁻¹
4934.1	Ba II $6^2S_{1/2}-6^2P_{1/2}$	10, 12	0.174	0.118
4579.7	Ba I $6s6p^2P_2-^412''$	10, 12, 17.5	0.100	0.098
4620.0	Ba I $6s6p^2P_0-6s8s^2S_1$	12	0.106	0.166
4691.6	Ba I $6s6p^2P_2-6p^2^2P_1$	10, 12, 22, 30	0.093	0.078
5519.1	Ba I $6s6p^2P_1-6s6d^3D_2$	12	0.078	0.045
5680.2	Ba I $6s5d^3D_2-^450''$	7.5, 22, 30		0.113
6110.8	Ba I $6s5d^3D_3-fd6p^2P_2$	17.5, 19.5, 30	0.055, 0.095	0.063

component which tends to obscure the detail of the structure due to the two odd isotopes.

EXPERIMENTAL

The barium spectrum was excited in a water-cooled hollow cathode discharge tube in an atmosphere of helium. The current used varied from one-tenth to one ampere on different plates. The spectrum was studied with a Fabry-Perot interferometer, with quartz plates. The plates were silvered by evaporation with an apparatus similar to that of Ritschl.¹¹ Various plate separations, from ten to thirty millimeters, were employed. The interferometer was used both with a large Littrow glass spectrograph and with a two-prism glass spectrograph.

Measurable hyperfine structure was found in a number of lines. Table I gives the results and shows, for each line, its classification, the plate separations used, and the separation of the components from the heavy central component due to the even isotopes. In each case, the result is the average of measurements from several plates.

DISCUSSION OF RESULTS

Since few of the barium hyperfine structure patterns are completely resolved and all are obscured by the heavy central component already mentioned, indirect methods must be used to determine the nuclear moment. Murakawa and Kruger, Gibbs, and Williams relied on the intensity ratios of the components of the 6^2S-6^2P lines of Ba II. This is, at best, a difficult and uncertain procedure and liable to errors due to plate background, plate calibration, and other experimental troubles which may

easily lead to false conclusions, since the intensity ratios for different spins differ but little. Thus Schütz, in his work on the intensity ratios of the hyperfine structure of the blue caesium doublet, at first reported $I=1\frac{1}{2}$ ¹² and in a later correction¹³ gave $4/2 \leq I \leq 7/2$ with $I=2\frac{1}{2}$ as probable, while the correct value is now known to be $I=3\frac{1}{2}$.¹⁴ Schüler and Jones do not explain their procedure but perhaps used the separations of the components of the Ba II resonance line $6^2S_{1/2}-6^2P_{1/2}$ from the center of gravity. This method involves an assumption as to the presence of isotopic shift and is hardly adequate as a sole criterion. In the present work an attempt has been made to bring to bear evidence from several points of view in order to strengthen the conclusion.

The most direct method of approach is that mentioned above—the separations of the components of the $^2S_{1/2}$ terms from their center of gravity. The combination of a $^2S_{1/2}$ term with a term of no structure or unresolved structure gives rise to a pattern of only two components for all values of I , since the multiplicity is $(2J+1)$, but the relative intensities and separations depend on I . The intensities are proportional to $2F+1$ and the distances from the center of gravity are thus inversely proportional to $2F+1$. Since the F values are $I+\frac{1}{2}$ and $I-\frac{1}{2}$, the distances must be proportional to I and $I+1$. The line $6^2S_{1/2}-6^2P_{1/2}$ is convenient for the test, since the structure of $6^2P_{1/2}$ is probably small and has never been resolved. If we assume no isotope shifts, and no effect from the $6^2P_{1/2}$ splitting, then the ratio of the distances of the hyperfine structure components should be equal to the ratio of I to $I+1$ and I may be computed.

¹² Schütz, Naturwiss. 19, 774 (1931).

¹³ Schütz, Naturwiss. 19, 1007 (1931).

¹⁴ Kopfermann, Zeits. f. Physik 73, 437 (1931).

¹¹ Ritschl, Zeits. f. Physik 69, 578 (1931).

The line, $\lambda 4554$ has been examined by several observers with the results listed in Table II. The evidence clearly favors $I=1\frac{1}{2}$ over any other odd multiple of $\frac{1}{2}$. The variation in values indicates the difficulty of measuring these patterns, which are not completely resolved from the central component.

In the above calculation we have taken simply the distances of the unresolved components from the central components. In analyzing their data, Kruger, Gibbs, and Williams assumed that the ${}^2P_{11}$ level had both a splitting of 0.033 cm^{-1} and an isotopic shift of 0.017 and concluded that, assuming no shift for ${}^2S_1$, its odd levels had separations of 0.113 and 0.159 . These separations would lead to a value of I , by the above method, of 2.45 . As a partial check, however, on this interpretation of ${}^2P_{11}$ we may consider the line, $6^2P_{11}-7^2S_1$, $\lambda 4899.9$, which was resolved by Murakawa. He assigned the splitting, $+0.043$ and -0.027 , to 7^2S_1 . The above method of calculation gives, then, $I/I+1=0.027/0.043$ or $I=1.68$, in good agreement with the calculations on 6^2S_1 , and in support of the idea of small isotope shift and small 6^2P_{11} splitting. The analysis of the $6^2P_{11}-7^2S_1$ may also be studied graphically. In Fig. 1, is shown at the right, above, the level scheme, and below, the resulting grouping of the components about the central components for Murakawa's data and assignment. In Fig. 1 left, is shown the level scheme and line appearance on the assumption by Kruger, Gibbs, and Williams of no 7^2S_1 splitting, and of splitting and isotope shift in 6^2P_{11} . In Fig. 1 center, is a combination of both Kruger's and Murakawa's idea. The former's value for 6^2P_{11} splitting is used while Murakawa's 7^2S_1 splitting is used but with a center of gravity corresponding to $I=2\frac{1}{2}$. It is clear that neither the left or central figures represent structures which are in accordance with Murakawa's data.

TABLE II. Nuclear moment of barium computed from $\lambda 4554$.

OBSERVER	$I/(I+1)$	I
Murakawa	$0.099/0.154=0.64$	1.80
Kruger <i>et al.</i>	$0.085/0.164=0.52$	1.07
Ritschl and Sawyer	$0.09/0.16=0.56$	1.30
Benson and Sawyer	$0.100/0.152=0.66$	1.92
Mean		$1.52 \pm 0.17 (\pm 2d/n^3)$

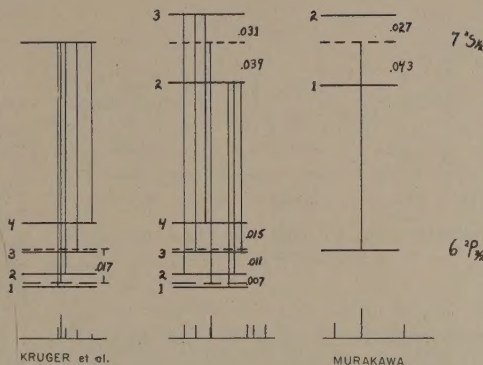


FIG. 1. Level scheme and line structure of $6^2P_{11}-7^2S_1$, 4899.9 . 1. According to Murakawa's data and assignment (right). 2. According to the Kruger, Gibbs, and Williams explanation of splitting and shift of the 6^2P_{11} term (left). 3. According to a combination of the two assignments (center).

Further evidence on the nuclear moment of barium may be obtained from the Ba I line $6s5d^3D_3-5d6p^3P_2$, $\lambda 6110.8$, which has been resolved into four components in this work, as well as by Murakawa and by Ritschl and Sawyer (unpublished material). The structure must be due to the ${}^3D_3$ term, for little structure would be expected in the ${}^3P_2$ term since it has no s electron. Furthermore, Murakawa found the same structure in $6s5d^3D_3-5d6p^3D_3$, $\lambda 6498.78$, which is beyond the present range. The term ${}^3D_3$ should have four components for $I=1\frac{1}{2}$, or six for $I=2\frac{1}{2}$, but, if the interval rule is obeyed, the spacing should be quite different in the two cases. Fig. 2 shows how the ${}^3D_3$ level would appear if I were $1\frac{1}{2}$ and if it were $2\frac{1}{2}$. Since the center of the pattern is obscured by the even component, the best approach is to consider the ratio of the spacing of the two components with smallest F to the over-all width. From the Landé interval rule, for $I=1\frac{1}{2}$, this is $5/21=0.238$, while for $I=2\frac{1}{2}$, it is $3/55=0.085$. If, in the latter case, the farthest component was not observed, the ratio of the next two components to the total width would be $5/31=0.155$. The observations give:

Murakawa	$0.037/0.156=0.238$
Ritschl and Sawyer	$0.032/0.147=0.218$
Benson and Sawyer	$0.038/0.156=0.244$

The data points definitely to $1\frac{1}{2}$ as the value of the nuclear moment, and it may be emphasized

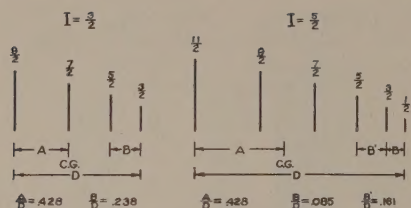


FIG. 2. Splitting of the $^3D_3$ term if $I=2\frac{1}{2}$ (right) and if $I=1\frac{1}{2}$ (left).

that the ratios used do not depend on the assumption that the isotope shift is zero. As proof that the outermost and weakest component was observed, we may consider the ratio of the separation of the strongest line from the center of gravity to the over-all width. The ratio is 0.428 both for $I=1\frac{1}{2}$ and $I=2\frac{1}{2}$, while, if the faintest component were missed, it would be 0.563 for $I=1\frac{1}{2}$ and 0.470 for $I=2\frac{1}{2}$. The measurements are:

Murakawa	0.060/0.156=0.348
Ritschl and Sawyer	0.057/0.147=0.388
Benson and Sawyer	0.063/0.156=0.404

The indication is that 0.428 is the ratio and that the isotope shift, if any, is small, since the amount of isotope shift required to bring the above ratios to exactly 0.428 is:

Murakawa	0.007
Ritschl and Sawyer	0.006
Benson and Sawyer	0.003

The conclusion is that the outermost component was observed, and the first ratios quoted give a very definite preference for $I=1\frac{1}{2}$ rather than $I=2\frac{1}{2}$, since the ratios are widely different in the two cases.

Further evidence from $\lambda 6110.8$ is given in Fig. 3, which shows a graphical comparison of the expected and observed structures for a $^3D_3$ term. The lines above the horizontal line show the experimental values obtained by Murakawa and by the authors, while the lines below show the expected structure, drawn with the same over-all width for $I=1\frac{1}{2}$ and for $I=2\frac{1}{2}$. It will be noted that the experimental results are in good agreement with $I=1\frac{1}{2}$. The fact that the values fall a little to the left of the calculated positions with respect to the center of gravity (placed at the central even component) indicates a possible small isotopic shift.

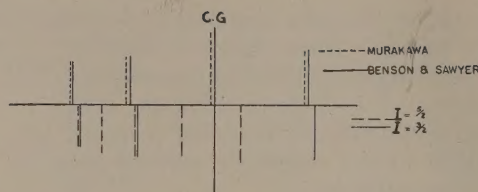


FIG. 3. The upper part shows the observed data of Murakawa and of Benson and Sawyer, for the line $6s5d^3D_3-5d6p^3P_2$, 6110.8. The lower part shows the expected structure, with the same over-all spread as observed, from a $^3D_3$ term if $I=1\frac{1}{2}$ and if $I=2\frac{1}{2}$.

Similarly the structure of the lines $\lambda 4691.6$ ($6s6p^3P_2-5d6d^3P_1$) and $\lambda 4579.7$ ($6s6p^3P_2-5d6d^1D_2$) is shown graphically in Fig. 4. Although the structure is slightly different, it may be safely assumed to arise essentially from the level 6^3P_2 . The structures below the lines in the graph are those for $^3P_2$ with the over-all widths adjusted to be the same as those of the observed structures. It will be seen that only the structure for $I=1\frac{1}{2}$ can be brought into harmony with the observations.

The Goudsmit and Bacher constant a_1^{12} has been calculated for several of the lines and comes out nearly a constant, both for $I=1\frac{1}{2}$ and for $I=2\frac{1}{2}$. The evidence is not directly in favor of either value of nuclear moment, but since, in two of the lines, the distance from center of gravity to the outer component was used, instead of over-all width, the agreement of the values means small isotope shifts. As we have seen, the assumption of small isotope shifts is in better accord with the correlation with $I=1\frac{1}{2}$ than with $I=2\frac{1}{2}$. The computed values for A and a_1 are given in Table III. It will be noted that the only considerable variations are in the ^3D terms. Similar deviations were noted by Goudsmit and

TABLE III. Computed values of A and a_1 for barium.

LINE	CONFIGURATION	EXP. TOTAL WIDTH	A FROM $I=1\frac{1}{2}$	A FROM $I=2\frac{1}{2}$	a_1 FROM $1\frac{1}{2}$	a_1 FROM $2\frac{1}{2}$
4620	$6s8s^3S_1$	0.274	0.068	0.045	0.100	0.067
4691	$6s6p^3P_2$	*	0.026	0.016	0.104	0.062
4579	$6s6p^3P_2$	0.198	0.026	0.016	0.106	0.066
6111	$6s5d^3D_3$	0.156	0.015	0.009	0.089	0.054
5680	$6s5d^3D_2$	*	0.038	0.023	0.152	0.090

* In the case of two of the lines above, the distance from the center of gravity to the strongest component, was measured and then from this value A was computed. In the other cases A was obtained from the over-all width.

Bacher in the ^3D terms of the analogous Tl II spectrum.

CONCLUSION

It has been assumed throughout the preceding discussion that the even isotopes have no spin and the line due to each of them falls undisplaced and unresolved at the center of gravity. These assumptions seem to be established within the limits of the resolution attained. The spectra of the two odd isotopes are also assumed to fall together and to have the same nuclear moment, as happens in most known similar cases. The evidence given on the basis of the separation of the components from the center of gravity, on the basis of the spacing, and on the basis of the patterns of the various lines, while not absolutely conclusive, when taken with the intensity measurements of Murakawa, seem to

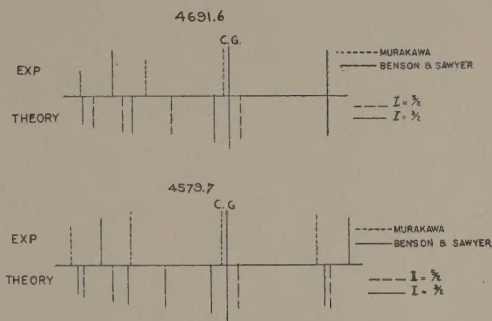


FIG. 4. The observed and expected structures are shown for $6s6p^3P_2-5d6d^3P_1$, 4691.6, and for $6s6p^3P_2-5d6d^1D_2$, 4579.7, on the assumption that the structure is due to the $^3P_2$ term. The calculated structures are given both for $I=1\frac{1}{2}$ and for $I=2\frac{1}{2}$ and are drawn to have the same over-all width as the observed patterns.

point quite convincingly to a value of $I=1\frac{1}{2}$ for the nuclear moment of the odd isotopes of barium.

